In Situ X-Ray Studies of MOCVD $Pb(Zr,Ti)O_3$ Growth

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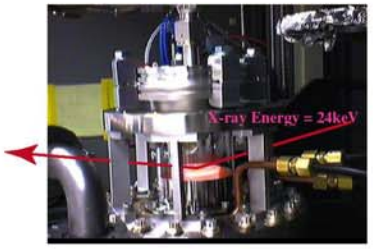
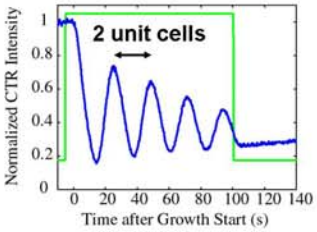
Motivation:

- Ferroelectric thin films hold great potential for a wide variety of applications (e.g., nonvolatile memories, sensors, actuators)
- Challenge:
  - ❖ Grow high quality homogeneous ultrathin films with controlled composition
    - ➢ Strain Development
    - ➢ Morphology Instability
    - ➢ Composition Variation

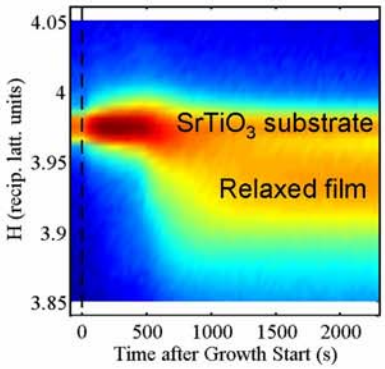
Approach:

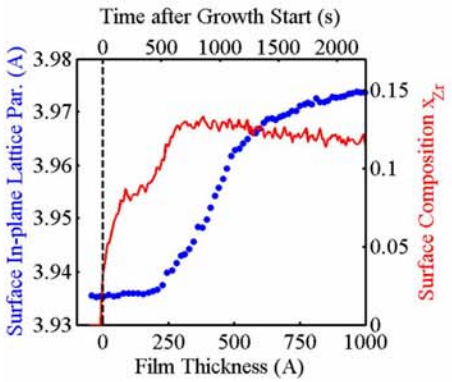
- Unique apparatus at APS 12ID-D allows in-situ monitoring of the growth of metalorganic chemical vapor deposition
- *In situ* synchrotron x-ray scattering to control growth and observe ferroelectric transition as well as strain state of heterostructures with well-defined thickness and boundary conditions
- *In situ* synchrotron total reflection x-ray fluorescence to monitor the evolution of surface composition with the depth resolution of 2-3 nm





Accomplishments:





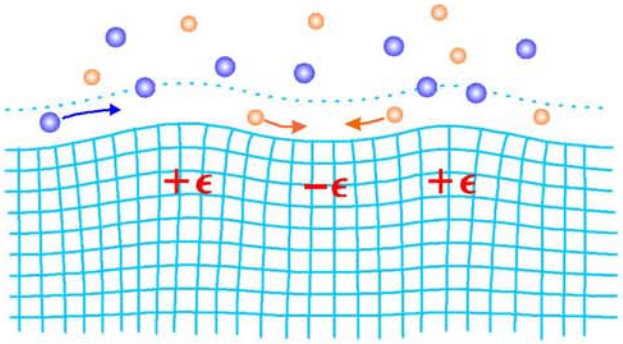
Observations:

- During coherent growth, Zr incorporation is limited.
- As the film relaxes, the Zr incorporation efficiency is limited; Zr composition increases.

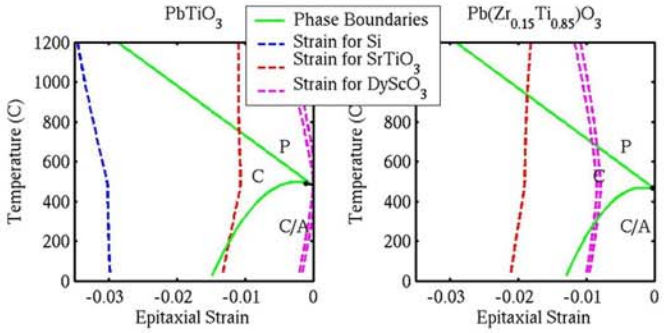
Impact:

- Strain state is intimately tied to the composition control during film growth.
- The coupling between strain and composition may influence the practical critical thickness limit for ferroelectricity in $Pb(Zr,Ti)O_3$.
- The optimization of both strain and composition is critical in designing future nanoscale devices.

Future Directions:



- Strain relaxation can drive surface instability and induce composition segregation. With CNM x-ray nanoprobe, can we monitor the lateral surface segregation in complex oxide oxide system?



Phase diagrams calculated for various epitaxial strains acheived by changing compositions of $Pb(Zr,Ti)O_3$ and substrate lattice parameter

- We will investigate the dependence of T_c on epitaxial strain by studying ferroelectrics with different stress-free lattice parameters (e.g. $Pb(Zr_xTi_{1-x})O_3$ on substrates such as $SrTiO_3$ and $DyScO_3$. Can we obtain ferroelectricity in films as thin as one unit cell?

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